

## Aerosol Radiative Forcing of Asian Continental Outflow

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Anthropogenic aerosols are a major uncertainty in climate prediction. Current estimates of the global annually averaged direct radiative forcing of sulfates, soot (black carbon), mineral dust, and biomass smoke range from  $-0.3$  to  $-1.0 \text{ W m}^{-2}$ , with an uncertainty factor of about two. To reduce this uncertainty via better characterization of aerosol properties, integrated field experiments have been conducted (for example, Smoke, Cloud, and Radiation-A (SCAR-A), Smoke, Cloud, and Radiation-Brazil (SCAR-B), Indian Ocean Experiment (INDOEX), Tropospheric Aerosol Radiative Forcing Observation Experiment (TARFOX), and South African Regional Science Initiative (SAFARI)). Presented here are aerosol radiative forcing data for the western Pacific Ocean region (not covered by any of the above-mentioned deployments) with simulations based on in situ measurements of particle characteristics in elevated Asian outflow.

As an illustration of the findings, figure 1 compares the clear-sky daily averaged aerosol forcing of Asian outflow (column 1) per unit optical depth with other types of forcings, namely biomass burning (ScB and Zam), forcing by dust (Ace2), forcing associated with urban-industrial pollution (Tfox, ScA, Indo), and forcing by enhanced stratospheric aerosol (Pin). Results are presented for solar broadband fluxes at the surface and at the top of the atmosphere, and for infrared broadband fluxes at the top of the atmosphere. It follows from the first figure that (1) the daily averaged forcing varies between  $-60$  and  $-100 \text{ W m}^{-2}$  irrespective of the geographic location of the measurements; (2) the averaged forcing at the top of the atmosphere is usually significantly

smaller than at the surface; and (3) infrared heating at the top of the atmosphere in Asian continental outflow is similar to that of Saharan dust.

In spite of only a relatively small change in the net flux at the top of the atmosphere, generally looked at as a measure of the climatic impact of aerosols, Asian outflow strongly reduces the solar radiation reaching the surface. Possible consequences are dynamical feedbacks (for example, suppressed convection, weakening of the hydrological cycle, and changes in local heating rates.) Heating rates of  $+10 \text{ kelvin/d}$  (K/d) (per unit mid-visible extinction) were calculated, mainly they were caused by sub-micron particles. Particles exceeding one micrometer contributed only about 30% to this rate due to reduced solar heating because of infrared cooling. This atmospheric heating by aerosols could cause low-level clouds to evaporate. This is an opposing effect to increase in cloud cover that can be attributable to more cloud condensation nuclei forming smaller drops at higher concentration.

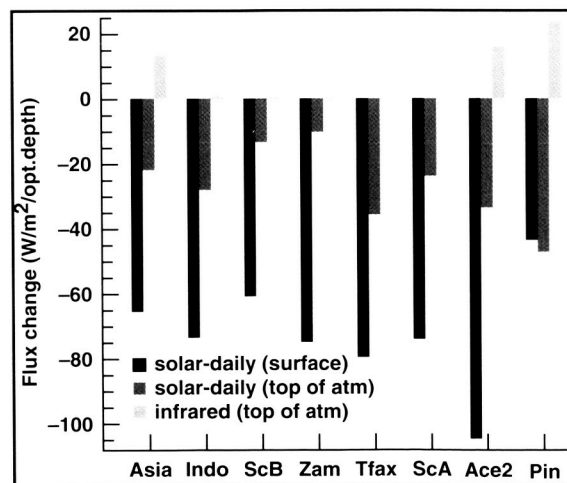


Fig. 1. Scaled aerosol direct forcing comparison.

Although stratospheric background aerosol is nonabsorbing, the particles injected by the Pinatubo volcanic eruption were large enough to cause infrared warming (Fig. 1). In the Arctic stratosphere, the existence of soot aerosol (arguably from aircraft) increases heating rates from a fraction of a degree per day to about two degrees per day, with possible implications to dynamics and chemical reaction rates.

The presence of clouds is critical for an assessment of aerosol forcing. In the presence of clouds at altitudes higher than the aerosol layer, the aerosol forcing is similar to a cloud-free scenario, albeit at a reduced rate. If the

clouds are below the aerosol layer, aerosols primarily reduce cloud-associated solar flux losses such that cooling is not just reduced but can change to warming. Thus the removal of lower-level clouds turned weak net flux gains (warming) into weak net-flux losses (cooling). S. Kinne of NASA Goddard Space Flight Center collaborated with the Ames investigators.

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## Analysis of SOLVE Observations of PSCs and Implications for the Evolution of the Arctic Vortex

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In-situ measurements of polar stratospheric clouds (PSCs) made from the ER-2 aircraft during the SAGE III Ozone Loss and Validation Experiment (SOLVE) have revealed new information about the composition and properties of these clouds. Model results have been compared with the ER-2 measurements in order to refine understanding of the cloud microphysics.

Measurements made by the Multiangle Aerosol Spectrometer Probe (MASP) reveal that most particles generally appear to be liquid-phase. Growth of these particles was observed at temperatures near 192 kelvin (K), consistent with the predicted behavior of ternary nitric acid/dihydrogen sulfate/water solutions ( $\text{HNO}_3/\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ ). Even at warmer temperatures, starting as air cools below 196 K, some swelling of the particles is apparent. The correlation of this growth with total available  $\text{HNO}_3$  indicates that  $\text{HNO}_3$  condensation is responsible. The observations have been compared with predictions of several ternary

solution models with generally good agreement. The presence of liquid-phase particles has implications for the long-term evolution of the Arctic winter. The widespread persistence of liquid particles constrains understanding of freezing processes, especially in air parcels that have experienced extensive denitrification. No significant differences in particle size distributions exist between highly denitrified and less denitrified air parcels, suggesting that very few particles are removed by the processes causing denitrification.

However, a very small fraction ( $<0.1\%$ ) of the particles did freeze during the winter, forming solid-phase  $\text{HNO}_3$ -containing particles that were observed both by the MASP and by the  $\text{NO}_y$  instrument. The presence of these frozen particles cannot be explained by current laboratory data on aerosol freezing, in which freezing occurs only at temperatures several degrees below the ice frost point. Alternative formation mechanisms, in particular homogeneous freezing above the ice frost point and